

Microstructural Characterization of Atomized UAl_x Powder for High-Density LEU Dispersion Target Fabrication

Sungchan Park, Ki Nam Kim, Jong Hwan Kim, Sunghwan Kim, Kyu Hong Lee, Yong Jin Jeong, Jong Man Park
Research Reactor Fuel Development Division, Korea Atomic Energy Research Institute, 111, Daedeok-daero
989beon-gil, Yuseong-gu, daejeon, Republic of Korea
E-mail: sungchan@kaeri.re.kr

1. Introduction

Tc-99m can be widely used in the radiopharmaceutical field owing to its short half-life and gamma radiation of 140 keV [1]. Tc-99m has been obtained from the decay of Mo-99, which is the fission production of U-235 in research reactors [2, 3]. The Korea Atomic Energy Research Institute (KAERI) has been developing high-density low enriched uranium (LEU) dispersion targets to improve the Mo-99 production efficiency of conventional LEU targets. Until now, UAl_x powder has been successfully fabricated through centrifugal atomization, and the results have already been reported [4]. For the improvement in Mo-99 production efficiency, however, it is essential to understand diverse phenomena such as the phase transformation of U-Al alloy, the formation of intermetallic compounds, defects, and metallurgical interactions. The objective of this study is to evaluate the microstructure in atomized U-Al alloys with varying Al composition in more detail.

2. Experimental Procedures

$U-xAl$ ($x= 0, 5, 10, 15, 20,$ and 25 wt.%) alloy powders were fabricated using a centrifugal atomization technique at KAERI. Before the induction melting process, U-Al mother alloy was arc-melted in an argon atmosphere to prevent the thermal shock. After charging the mother alloy into a ZrO_2 , the melting chamber was induction-heated in Ar to a temperature of $300^\circ C$ higher than the melting point of intermetallic compounds to increase the fluidity of molten metal, which is then fed onto a rotating graphite. Subsequently, molten metal droplets were spread from the disk toward the chamber wall and cooled under Ar. Fig. 1 shows a schematic processing of centrifugal atomization for UAl_x powder.

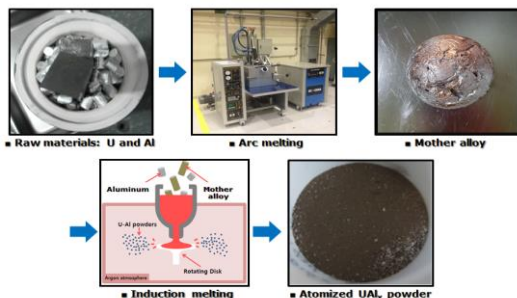
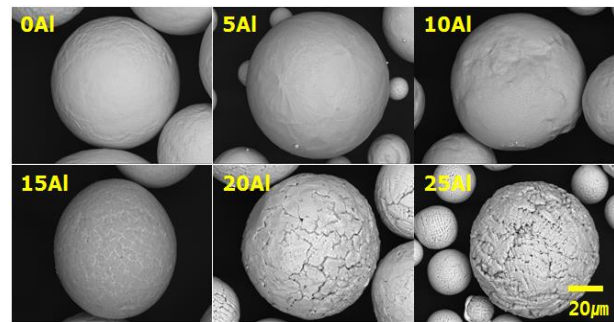


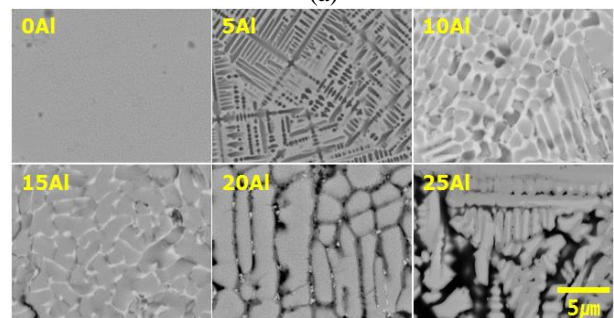
Fig. 1. A schematic processing of centrifugal atomization

3. Results and Discussion

$U-xAl$ ($x= 0, 5, 10, 15, 20,$ and 25 wt.%) alloy powders were investigated using SEM, EDX, and XRD to evaluate the microstructure according to the Al composition. In the atomized powder, it was observed that spherical particles with an average size of $70 \mu m$ were obtained, as shown in Fig. 2(a). When the Al composition was more than 20 wt.%, remarkable cracks occurred on the surfaces of the particle owing to solidification shrinkage of the liquid phase, and this change in the morphology can be explained through a phase diagram [5]. Additionally, the surface morphology depends on the Al composition because of the fractions of intermetallic compounds such as UAl_2 and UAl_3 . Fig. 2(b) shows a cross-section micrograph of $U-xAl$ alloy particles a higher magnification and it can clearly distinguish the matrix, boundary, and dendrite. As the Al composition increased, the dendrite microstructure was observed in the matrix owing to constitutional supercooling during the cooling process.



(a)



(b)

Fig. 2. SEM images collected on $U-xAl$ ($x= 0, 5, 10, 15, 20, 25$ wt.%) particles. (a) surface morphology, (b) cross-section micrograph.

Regarding the U-Al intermetallic compounds, it can be predicted from Fig. 3, even if a centrifugal atomization is a thermodynamic non-equilibrium due to rapid solidification process. Therefore, the three possible uranium aluminides can be UAl_2 , UAl_3 , and $U_{0.9}Al_4$, but the $U_{0.9}Al_4$ was not formed because it was fabricated up to 25 wt.% Al.

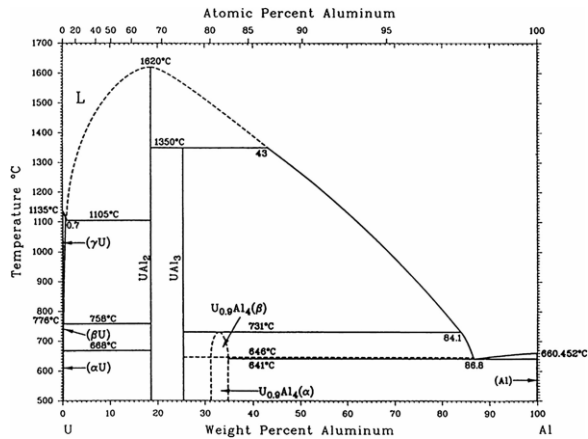


Fig. 3. Phase diagram of U-Al system [5]

As shown in Fig. 4, the EDX results obtained from a representative U-10 wt.%Al particle also confirmed that the bright-colored matrix was U, and the dark phase was U-Al intermetallic compounds. Based on a U-Al binary phase diagram with fraction, the intermetallic compound and matrix were identified as UAl_2 and α -U, respectively.

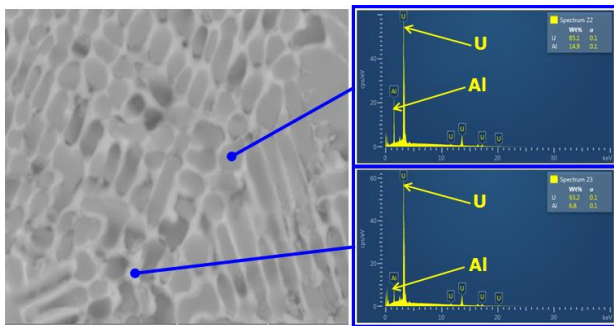


Fig. 4. SEM image and EDX spectrum of U-10 wt.% Al particles.

XRD was used to determine the crystal structure and phase information of metallic U and UAl_x intermetallics. Fig. 5 shows that UAl_x consisted of compounds such as UAl_2 , and UAl_3 . In the U-Al alloy with an Al composition of less than 15 wt.%, peaks of α -U and UAl_2 appeared. When the Al composition was 20 wt.% to 25 wt.%, the peak of the UAl_3 phase was observed in the XRD data. Regarding the crystal structure of UAl_x , the α -U phase is an orthorhombic system, and the phase

of UAl_2 and UAl_3 are a cubic system [6, 7]. The crystallographic structure and physical properties of the uranium aluminides are shown in Table 1. These results are in agreement with the results obtained through the SEM and EDX.

Table 1. Structure and physical properties of three uranium aluminides [7]

	UAl_2	UAl_3	$U_{0.9}Al_4$
Crystal structure (space group)	Cubic ($Fd\bar{3}m$)	Cubic ($Pm\bar{3}m$)	Orthorhombic ($Imma$)
Lattice parameter (nm)	$a = 0.7766$	$a = 0.4287$	$a = 0.4397$ $b = 0.6251$ $c = 1.3714$
wt% U	81.52	74.63	64.2–66.3
Density (g/cm^3)	8.14	6.8	5.6–5.7
Melting point (K)	1,893	1,623	1,004

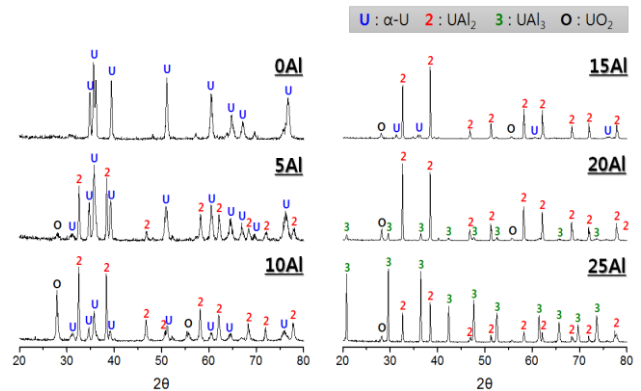


Fig. 5. XRD data collected on U-xAl ($x = 0, 5, 10, 15, 20, 25$ wt.%)

4. Conclusions

KAERI has been developing a high-density LEU dispersion target technology using atomized U-Al alloy powder to improve the Mo-99 production efficiency. In this study, U-Al alloys with Al composition varying from 0 wt.% to 25wt.% were fabricated using a centrifugal atomization, and were characterized through SEM, EDX, and XRD.

Considering the analysis results, we can conclude that the fabricated U-Al alloy powder was obtained with an average size of $70 \mu m$ and a spherical morphology, and intermetallic compounds such as α -U, UAl_2 , and UAl_3 , with a dendritic structure, and microstructural changes were observed according to the Al composition. Thus, we confirmed crystallographic structure and physical properties of uranium aluminides. As a result, it is possible to understand the microstructural characteristics of the U-Al alloy, which can be used as important evidence for the composition design and fabrication process for a high-density LEU dispersion target.

Acknowledgement

This research was supported by Nuclear Energy and Safety Program of the National Research Foundation of Korea (NRF) grant funded by the Korea government (MSIT) (No. 2017M2A8A1016345).

REFERENCES

- [1] National Academies of Sciences, Engineering, and Medicine, Molybdenum-99 for Medical Imaging, National Academies Press, Washington, DC, pp. 26, 2016
- [2] SC. Marck, The options for the future production of the medical isotope ⁹⁹Mo, European Journal of Nuclear Medicine and Molecular Imaging, Vol. 37, pp. 1817, 2010
- [3] A. Mushtaq, Reactors are indispensable for radioisotope production, Annals of Nuclear Medicine, Vol. 24, pp. 759, 2010
- [4] H. J. Ryu, Development of high-density U/Al dispersion plates for Mo-99 production using atomized uranium powder, Nuclear Engineering and Technology, Vol. 45, pp. 979, 2013
- [5] T. Massalski, Binary alloy phase diagrams, American society for metals, Metals park, 1986
- [6] M. E. Kassner, Evaluation and thermodynamic analysis of phase equilibria in the U-Al system, Journal of Nuclear Materials, Vol. 167, pp. 160, 1989
- [7] H. J. Ryu, Metallurgical considerations for the fabrication of low-enriched uranium dispersion targets with a high density for ⁹⁹Mo production, Journal Radioanalytical Nuclear and Chemistry, Vol. 305, pp. 31, 2015